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The Self Attenuation Correction for Holdup
Measurements, an Historical Perspective

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The Self Attenuation Correction for Holdup Measurements, an Historical Perspective

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Abstract

Self attenuation has historically caused both conceptual as well as measurement problems. The purpose of this paper is to eliminate some of the historical confusion by reviewing the mathematical basis and by comparing several methods of correcting for self attenuation focusing on transmission as a central concept.

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1 Introduction

Self attenuation has historically caused both conceptual as well as measurement problems. The purpose of this paper is to eliminate some of the historical confusion by reviewing the mathematical basis and by comparing several methods of correcting for self attenuation focusing on transmission as a central concept. Some of the historical misconceptions will be reviewed.

2 Mathematical basis for the self-attenuation correction

The conceptual problem for the self-attenuation correction occurs because matter is divided into two categories for the purpose of analytical radiation transport. The first category of matter attenuates gamma rays but does not produce gamma rays. The second category of matter both produces and attenuates gamma rays.

Container and equipment walls are typical of the first category of matter. The change in the number of gamma rays through a differential thickness of this matter is negative and proportional to the number of gamma rays and a differential thickness of material. For the one dimensional case

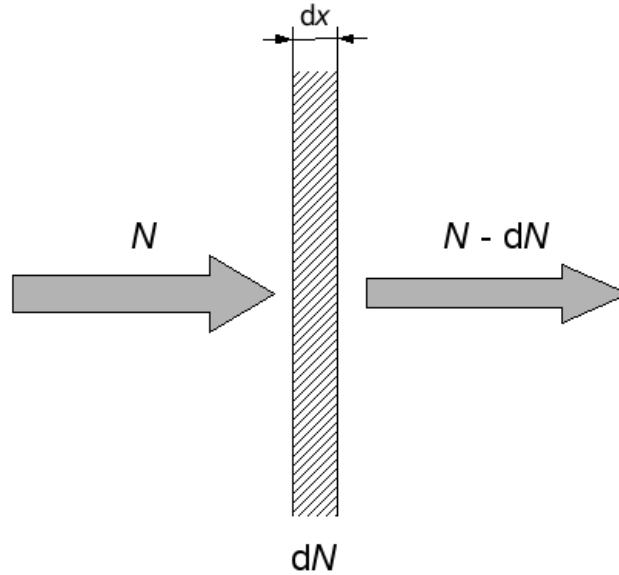


Figure 1: Change in number of gamma rays through a differential thickness dx of matter.

$$dN = -kNdx. \quad (1)$$

The solution to this differential equation is the familiar

$$N = N_0 e^{-kx} \quad (2)$$

where N_0 is the initial condition and $k = \mu\rho$ is the product of the mass attenuation coefficient and density of the matter through which the gamma rays pass.

A transmission T through a thickness x of this material can be defined as

$$T = \frac{N}{N_0} = e^{-kx}. \quad (3)$$

The second category of matter attenuates as before with a constant of k_2 but also increases the radiation in proportion to the thickness of material

$$dN = k_1 dx - k_2 N dx. \quad (4)$$

The solution to this differential equation is the familiar

$$N = \frac{k_1}{k_2}(1 - e^{-k_2 x}). \quad (5)$$

The second category of matter causes confusion because it is a mathematical contrivance. It is applied to whatever mixture of matter that is convenient for the analysis. For holdup measurements of uranium, the quantity of ^{235}U is determined from the detection of 186keV gamma rays. The matter is usually defined as the ^{235}U along with any other matter with which it is more or less homogeneously mixed. The matter other than uranium is often referred to as the matrix. For uranium metal the matter would include the ^{235}U along with the ^{238}U and other isotopes. For uranium oxide, it would include the uranium isotopes and oxygen. For an aqueous solution, it would include the uranium as well as solvent. All of these examples easily meet the homogenous mixture criteria.

The mathematical contrivance is often extended to less homogenous mixtures such as contaminated combustibles or a HEPA filter loaded with uranium oxide and other dirt.[1] In the case of the HEPA filter, the filter paper, paper separators, uranium oxide and dirt would be treated as a homogenous mixture. This treatment is approximately true and makes the analysis possible.

The attenuation constant k_2 can be related to physical properties of the mixture. The mixture will have a bulk density ρ_m , a mass fraction, and a mass attenuation coefficient for the various components f_i and μ_i . The bulk attenuation constant is defined as $k_2 = \mu\rho = \sum_i f_i \mu_i \rho_m$. It is convenient to define the mass attenuation coefficient of the mixture as $\mu_m = \sum_i f_i \mu_i$.

A self attenuation correction factor can be developed from the transmission as defined by Equation (3). The number of gamma rays from the matter would be $N' = k_1 x$ if there were no attenuation. Therefore the correction factor is the ratio of N' and N .

$$CF_{self} = \frac{N'}{N} = \frac{k_2 x}{(1 - e^{-k_2 x})} = \frac{-\ln(T)}{1 - T}. \quad (6)$$

Equation (6) is the basis for the self attenuation correction. Because it is one dimensional, both a far field and narrow field of view are implied.[2] Furthermore, the material is assumed to be homogenous.

3 Self attenuation correction from a measured transmission

The transmission T in Equation (6) can either be measured, or it can be calculated. When measured, the transmission is the ratio of the attenuated detection rate to the unattenuated detection rate. Typically a transmission source is measured at the correct source-to-detector distance for the unattenuated detection rate R_U . This measurement is shown in Figure 2a. The material being measured will be enclosed in a container. The effects of the container need to be considered. Therefore the transmission through the container needs to be measured. To do this, the empty container is interposed between the source and detector as shown in Figure 2b. The count rate observed is R_{Tc} . Similarly, the material being measured is included. This measurement represents both the transmitted gamma rays and gamma rays from the object itself R_T . In a fourth measurement, the transmission source is removed and the object is measured alone for R_0 . Each of these configurations is shown in Figure 2.

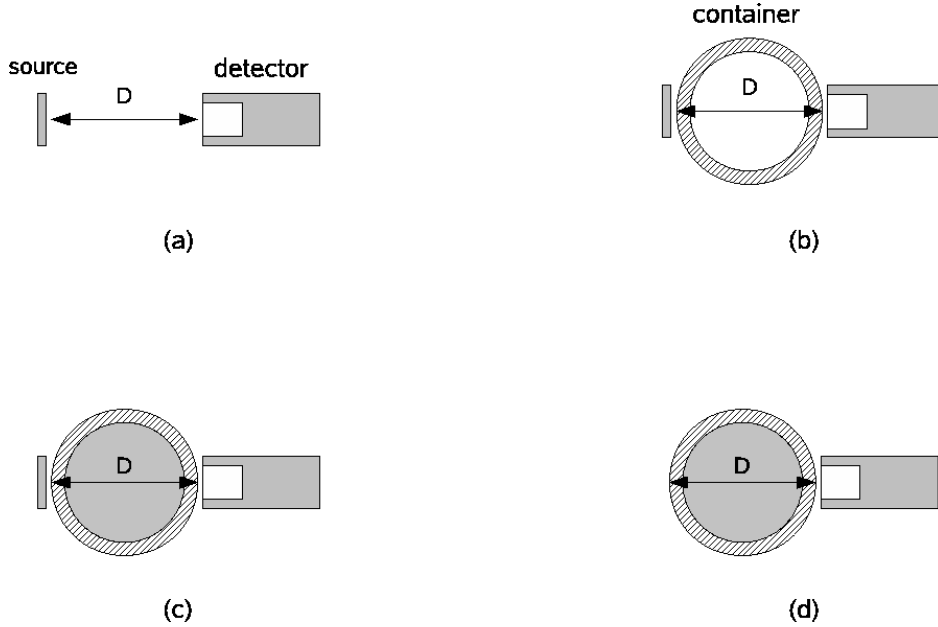


Figure 2: Measured transmission.

The measured transmission, with the effects of the container removed, is

$$T = \frac{R_T \frac{R_U}{R_{Tc}} - R_0 \sqrt{\frac{R_U}{R_{Tc}}}}{R_U}. \quad (7)$$

The transmission is a physical quantity. Typically the transmission is measured through a small area of the unknown material. When this transmission is used to correct for self

attenuation, there is an assumption that the measured transmission is representative of the bulk of the material. Furthermore when Equation (6) is used, there is an assumption of homogeneity.

The object being measured in Figure 2 is a cylinder. The transmission is measured through the full diameter D of the cylinder. A distributed transmission source would pass through various path lengths of the cylinder, none greater than D . In a far field approximation, all of these path lengths would be parallel. The average of these parallel path lengths is

$$\bar{D} = \frac{A}{D} = \frac{\pi}{4}D. \quad (8)$$

Therefore the self-attenuation correction factor for a cylinder is sometimes stated as

$$CF_{self} = \frac{k_2 \frac{\pi}{4}D}{(1 - e^{-k_2 \frac{\pi}{4}D})} = \frac{-\ln(T^{\frac{\pi}{4}})}{1 - T^{\frac{\pi}{4}}}. \quad (9)$$

The use of this average path length assumes both infinite far field and is restricted to two dimensions. Other values of the average path length have been proposed.[3]

4 Historical confusion in the application of the self attenuation correction

From the beginning it was known that self attenuation was a problem for holdup measurements.[4] In the first published demonstration of the generalized geometry holdup (GGH) method, the following correction for self attenuation was used:

$$CF_{self} = \frac{\mu(\rho x)_{measured}}{1 - e^{-\mu(\rho x)_{measured}}}. \quad (10)$$

It was recognized by holdup measurement practitioners that this correction factor underestimated the self-attenuation correction. There are at least three problems with Equation (10). First, it is the actual density thickness, corrected for the effects of self attenuation, not a “measured” density thickness that belongs in the equation. Second, it is the bulk density thickness of the mixture that is needed. Only the ^{235}U isotope is measured, but all the isotopes and other components of the mixture attenuate the gamma rays. All of these components need to be included. Finally, this equation implies a uniform distribution of the mixture which represents the distribution producing the minimum self attenuation.[5] Equation (6) is a more correct formula for the self-attenuation correction. However, there is still an assumption of homogeneity in this equation.

An additional problem with the self attenuation correction as it is applied to this day is that background is reduced for attenuation by equipment walls before it is subtracted but not for the attenuation of the uranium bearing material. This problem is treated in a separate paper.[6]

Recently Equation (11) was solved for the corrected density thickness which resolved the first problem with the equation.[7] However, the measured density thickness was not converted from the density thickness of ^{235}U to the density thickness of the entire mixture, leaving the second problem unresolved.¹

5 Self attenuation correction from a calculated transmission

Equation (6) can also be used with a calculated transmission. It is important to remember that $k = \mu_m \rho_m = \sum_i f_i \mu_i \rho_m$ where all of the components of the mixture are included. Substituting the calculated transmission into Equation (6) the self-attenuation correction factor can be used to convert a measured density thickness $(\rho x)_{measured}$ into a corrected density thickness $(\rho x)_{true}$:

$$(\rho x)_{true} = \frac{\mu_m \rho_m x_m}{(1 - e^{-\mu_m \rho_m x_m})} (\rho x)_{measured}. \quad (11)$$

Sometimes $\mu_m \rho_m x_m$ is known. Density can be determined from weight and volume. For $\mu_m \rho_m x_m$ to be known, the uranium contribution to attenuation must be insignificant since it is by definition an unknown.

6 Self transmission approach

Typically $\mu_m \rho_m x_m$ is not known. In fact one of the components to be measured is the ^{235}U component of the mixture. Equation (6) was traditionally applied by approximating $\rho_m x_m$ with $(\rho x)_{measured}$ as in Equation (10). However this substitution is simply incorrect. What is actually measured is the density thickness of ^{235}U . The density thickness of an individual component is related to the total density thickness of the mixture by

$$(\rho x)_i = f_i \rho_m x_m \quad (12)$$

¹See Reference [7] p. 20 Paragraph V.B. Although the mass-attenuation coefficient for the mixture is used, it is the areal density of the actinide rather than total mixture that is used.

Therefore to convert to the total density thickness of the mixture $\rho_m x_m = \frac{(\rho x)_i}{f_i}$. To convert the measured density thickness of ^{235}U to the total density thickness $(\rho_m x_m)_{\text{measured}} = \frac{(\rho x)_{\text{measured}}}{f_{U235}}$. Frequently the fraction of ^{235}U to material is broken up into two components, enrichment ϵ and grams U per gram f_U , $f_{U235} = \epsilon f_U$.

Frequently the fraction of uranium in a mixture remains constant with the quantity of the mixture varying. An example would be a holdup deposit consisting of a uranium compound such as U_3O_8 . Under these conditions, Equation (11) can be solved. To avoid any confusion, Equation (11) should be rewritten to reflect the actual meaning of $(\rho x)_{\text{measured}}$:

$$(\rho_m x_m) = \frac{\mu_m(\rho_m x_m)}{(1 - e^{-\mu_m(\rho_m x_m)})} \frac{(\rho x)_{\text{measured}}}{f_{U235}}. \quad (13)$$

Equation (13) can be easily solved for $(\rho_m x_m)$. The solution is

$$(\rho_m x_m) = \frac{-\ln \left[1 - \frac{\mu_m(\rho x)_{\text{measured}}}{f_{U235}} \right]}{\mu_m}. \quad (14)$$

The density thickness of material $(\rho_m x_m)$ can be converted to a density thickness of ^{235}U by multiplying the ^{235}U fraction f_{U235} . With some rearrangement Equation (14) becomes

$$(\rho x)_{U235} = f_{U235}(\rho_m x_m) = -\ln \left[1 - \frac{\mu_m}{f_{U235}}(\rho x)_{\text{measured}} \right] \frac{f_{U235}}{\mu_m}. \quad (15)$$

The factor $\frac{f_{U235}}{\mu_m}$ has been called the “normalizing mass attenuation coefficient.” [8] It can be calculated as

$$\frac{f_{U235}}{\mu_m} = \frac{f_{U235}}{f_{U235}\mu_U + \sum_i f_i\mu_i} \quad (16)$$

where the summation is understood to include all of the components of the mixture other than uranium. Mass attenuation coefficients of elements typically mixed with uranium in either a compound or mixture are listed in Table 1. The mass attenuation coefficient of uranium is more than a factor of ten greater than the others listed. Furthermore the mass attenuation coefficients of the other elements are very similar. Because of these factors, the exact composition of the mixture is not needed to determine the normalizing mass attenuation coefficient. The following simplification can be used:

$$\frac{f_{U235}}{\mu_m} = \frac{\epsilon f_U}{1.456 f_U + 0.125(1 - f_U)} \quad (17)$$

Table 1: Mass attenuation coefficients for common elements at 186 keV.

Element	Z	Mass Attenuation (cm^2/g)
H	1	0.2485
C	6	0.1248
N	7	0.1249
O	8	0.125
F	9	0.1185
Al	13	0.1219
Fe	26	0.144
U	92	1.456

The reason that this approach is called the self-transmission approach is because the argument of the natural logarithm in Equation (15) is the transmission T_m . It should also be noted that Equation (15) is valid only if there is no background.

For completeness, the case with background will be stated here. For a complete discussion see Reference [6]. It can be shown that the transmission with background can be inferred from the equipment corrected count rate R and the measured background B_0 as

$$T_m = \frac{\mu_m k_a R - f_{235U}}{\mu_m k_a B_0 - f_{235U}}. \quad (18)$$

Using this transmission, Equation (15) becomes

$$(\rho x)_{U235} = -\ln [T_m] \frac{f_{U235}}{\mu_m}. \quad (19)$$

Equations (15) and (19) are equivalent when $B_0 = 0$ and the substitution $(\rho x)_{measured} = k_a R$ is made.

7 Calibration approach for variable mass fraction of uranium

In some cases the fraction of uranium in a mixture is a variable. An example is the measurement of the uranium concentration in a solution.[8] When this occurs, Equation (13) cannot be solved for the corrected density thickness. Instead, Equation (13) can be rewritten and

the measured density thickness $(\rho x)_{measured}$, or even the count rate $R = \frac{(\rho x)_{measured}}{k_a}$, can be plotted as a function of the true density thickness:

$$R = \frac{\epsilon f_U [1 - e^{-\mu_m \rho_m x_m}]}{k_a \mu_m}. \quad (20)$$

The true density thickness $\mu_m \rho_m$ is the independent variable. It can be calculated from the fraction of uranium as follows:

$$\mu_m \rho_m = \sum_i f_i \mu_i \rho_m = [\mu_U f_U + \mu_s (1 - f_U)] \rho_m(f_U). \quad (21)$$

Substituting into Equation (20) results in

$$R = \frac{\epsilon f_U [1 - e^{-[\mu_U f_U + \mu_s (1 - f_U)] \rho_m(f_U)}]}{k_a [\mu_U f_U + \mu_s (1 - f_U)] \rho_m(f_U)}. \quad (22)$$

where R is the observed count rate, density as a function of the uranium concentration $\rho_m(f_U)$, and $\mu_s = \frac{\sum_i f_i \mu_i}{1 - f_U}$ is the mass attenuation coefficient of the solvent or matrix. It is understood for μ_s that i includes all of the components of the mixture except the uranium. The measured density thickness is related to the detector count rate by $(\rho x)_{measured} = k_a R$.

Equation (22) is solved by swapping the dependent and independent variables and fitting a polynomial to the plot of either f_U or ϵf_U as the dependent variable and R as the independent variable. An example of such a plot along with the polynomial fit is shown in Figure 3 where the concentration of ^{235}U is plotted as a function of measured count rate. The polynomial is $\epsilon f_U = 3.809 \times 10^{-11} R^3 - 4.106 \times 10^{-8} R^2 + 2.329 \times 10^{-4} R$.

There is a great deal of flexibility using the calibration approach. In this case the calibration was based on the analytical formula, Equation (22). Another approach might be to generate the calibration points using a Monte Carlo model of the holdup deposit. A traditional calibration results if physical models are substituted for the analytical formula or Monte Carlo model.

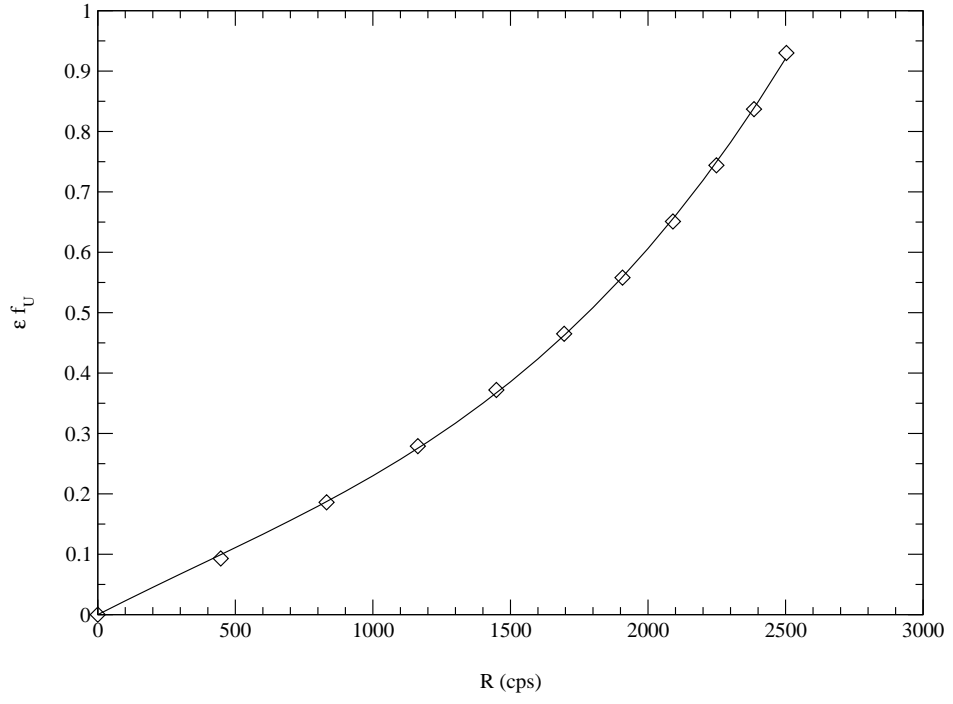


Figure 3: Example analytical calibration with the polynomial fit of $\epsilon f_U = 3.809 \times 10^{-11} R^3 - 4.106 \times 10^{-8} R^2 + 2.329 \times 10^{-4} R$.

8 Conclusion

The fundamental concepts for a self-attenuation correction have been reviewed. When the uranium and the matrix material are homogeneously mixed, the self-attenuation correction can be characterized in terms of the transmission. Several methods of determining the correction have been described and are summarized in Table 2.

Table 2: Summary of methods to determine the self-attenuation correction.

Method	Required Data	Usage	Equation
Measured transmission	-	An assumption of homogeneity is made when the measured transmission is used with Equation (6).	$CF = \frac{-\ln T}{1-T}$
Calculated transmission	μ_m, ρ_m, x_m	Appropriate when attenuation from uranium is insignificant. Usually weight, volume and composition are known. Equation (11)	$CF = \frac{\mu_m \rho_m x_m}{1 - \exp^{-\mu_m \rho_m x_m}}$
Self transmission	$\mu_m, \epsilon f_U$	Appropriate when f_U and μ_m are known and $\rho_m x_m$ varies. Equation (15) or (19)	$(\rho x)_{U235} = -\ln [T_m] \frac{f_{U235}}{\mu_m},$ $T_m = \frac{\mu_m k_a R - f_{235U}}{\mu_m k_a B_0 - f_{235U}}$
Calibration	$\mu_s, \epsilon, \rho_m(f_U)$	Typically the concentration of uranium in a known solution or matrix is measured. Equation (22)	$R = \frac{\epsilon f_U [1 - \exp^{-\mu_m \rho_m x_m}]}{K_a \mu_m}$

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